Rebuilding sources of linear tracers after atmospheric concentration concentration

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Atmos. Chem. Phys. Discuss., 3, 3171–3203, 2003
www.atmos-chem-phys.org/acpd/3/3171/

Atmospheric Chemistry and Physics Discussions

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Received: 11 March 2003 – Accepted: 12 June 2003 – Published: 19 June 2003

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Abstract

The identification of widespread sources of passive tracers out of atmospheric concentration measurements has become an important challenge of modern meteorology. The paper proposes some mathematical tracks to address this reconstruction of the complex space-time geometry of the sources. The methods are based upon the use of retroplumes. The inverse problem is addressed in a deterministic non statistical frame. The information obtained by local measurements is spread by introducing the concept of illumination. The constraint that the source to be rebuilt is non negative is also addressed. The experimental source ETEX1 is rebuilt in order to evaluate an impulse response of the algorithms.

1. Introduction

In a previous paper (Issartel and Baverel, 2003) we investigated the localisation of a point source after positive tracer concentration measurements. The situation corresponds to many anthropic pollutants accidentally released to the atmosphere from industrial installations having a fixed position at ground level. The method was based upon an interpretation of the adjoint transport equation as an inverse transport equation which enabled to address a Eulerian version of backtracking. This interpretation led to a very cost-effective calculation of the inverse or adjoint plumes related to the measurements, and called 'retroplumes'. The same theoretical elements are used here to investigate another type of sources. Such tracers as carbon dioxide, carbon monoxide, methane, are emitted on wide areas. In particular Bousquet et al. (2000) working with carbon dioxide have shown that methodological and physical angles must be jointly investigated. The source may vary in space and time, it may even become negative, a sink, where the tracer is absorbed.

The paper is based on an interpretation of the measurement operation as a scalar product, a point of view classically introducing the adjoint equations for the sensibility of the detectors and developed with various wordings by Pudykiewicz (1998), Enting (2000), Penenko and Baklanov (2001). Uliasz and Pielke (1991) introduced furthermore in this adjoint context the idea of inverse transport: the air sampled by the detector has originated from somewhere. A presentation of inverse transport has been proposed independently of adjoint techniques by Hourdin and Issartel (2000). The adjoint and inverse ideas were later described in (Issartel and Baverel, 2003) as different though equivalent. This context is reminded in Sect. 2. In Sect. 3 is developed the consequence of the measurements being regarded as scalar products: the source may be estimated by an orthogonal projection. This leads in fact to a least square estimation meeting the classical problem of data assimilation: how irrigating the whole system with a rare local information? In Sect. 4, instead of addressing this problem in terms of the statistical properties of the system, usually the statistical connections between its parts, it is addressed in terms of a deterministic property of the set of measurements, the 'éclairement' or 'illumination' describing which parts of the system are well or badly seen or not seen at all. The possibility of managing the constraint that the estimated source should everywhere be non negative is investigated in Sect. 5. This will enable to establish that the method is mathematically relevant, but that the physical relevance of the estimation may rely, besides the measurements, on independent additional physical constraints such as positivity. The stability of the estimation with respect to noises, biases and computational errors is investigated in Sect. 6.

The methods proposed in this paper are compared in Sects. 7 and 8 with the data collected during the first ETEX experiment. The experimental source of ETEX1 is a point source in space and, with a 12 h duration, almost also in time. Some people may consider it is not natural applying to ETEX1 methods devoted to widespread sources. Our purpose is not here to obtain an extensive information about the source known to be a point source. Rather, we want to determine, like Seibert (2001), the reliability of a method, and the resolution with which a source is seen in order to determine which smooth enough sources could be well seen. We want to obtain something like the 'impulse response' of an algorithm and it is necessary to use such a point source as

ETEX1 for that.

Sections 2 and 3 bear ideas going throughout the paper. The other sections are chained logically but the mathematical details are totally different and disconnected.

2. The measurement product

We investigate the possibility of identifying the source of a linear tracer out of concentration measurements. The unknown source is described throughout the atmosphere as a positive or negative rate of release $\sigma(\boldsymbol{x}, t)$ at position x and time t in unit amount of tracer per unit mass of ambient air and unit time. This source linearly generates a concentration $\chi(\boldsymbol{x}, t)$ in unit amount of tracer per unit mass of air (mass mixing ratio). The link between χ and σ stemming from a linear transport equation complemented with adequate boundary conditions may be described by a linear operator: $\chi = \mathcal{L}(\sigma)$.

The available tracer measurements μ_i, $i = 1, .., n$, correspond to the analysis of air samples taken at various positions and dates. Sampling functions $\pi_i(\boldsymbol{x}, t)$ describing where and when the samples have been taken are introduced in such a way that the measurements read as:

$$\mu_i = \int_{\Omega\times\mathrm{T}} \rho(\boldsymbol{x}, t)\ \chi(\boldsymbol{x}, t)\ \pi_i(\boldsymbol{x}, t)\ d\boldsymbol{x}\ dt \tag{1}$$

The integration is over the atmosphere Ω and the time domain T; ρ is the air density.

The unit of the π_i is a delicate though simple matter, it depends on the unit given to the measurements. Fundamentally the μ_i are amounts of tracer and the π_i are in inverse unit of time (in fact in unit mass of air sampled per unit mass of ambient air and per unit time). Nevertheless the measurements are generally presented as concentrations per unit mass of air or per unit volume. This means that the sampling functions are normalised in such a way that $\int \rho\ \pi_i\ d\boldsymbol{x}\ dt = 1$ or $\int \pi_i\ d\boldsymbol{x}\ dt = 1$, respectively, which implies a division by the total mass or volume of the air sample.

The measurement operates as a scalar product $\mu_i = \mu(\chi, \pi_i)$ of the tracer concentration and sampling functions. It is therefore possible to consider the adjoint operator

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$\mathcal{L}^*$ and the retroplumes $r_i = \mathcal{L}^*(\pi_i)$ and to rewrite the measurements $\mu(\mathcal{L}(\sigma), \pi_i) = \mu(\sigma, \mathcal{L}^*(\pi_i))$ as:

$$\mu_i = \int_{\Omega \times T} \rho(\boldsymbol{x}, t)\; r_i(\boldsymbol{x}, t)\; \sigma(\boldsymbol{x}, t)\; d\boldsymbol{x}\; dt \tag{2}$$

The adjoint calculation of the r_i is easy. In the case of a tracer passively transported by the air, with possibly some linear decay, the r_i may be interpreted as retroplumes. They describe the concentration among the ambient air of the air to be sampled, according to the π_i, before it was sampled. We stress that in this paper the convention is that *all detector functions are normalised with respect to the mass of the samples*, $\int \rho\, \pi_i\, d\boldsymbol{x}\, dt = 1$. The r_i are accordingly unitless mixing ratios.

The interpretation of the adjoint transport operator $\mathcal{L}^*$ as a backward transport operator requires an adequate choice of the conventions, mainly a system of units, for the source, concentration and sampling functions in order to reach the analytic form (1) of the measurement product. In the case of passive advection-diffusion $\mathcal{L}$ and $\mathcal{L}^*$ stand for the following forward and backward equations to be complemented with boundary conditions. The equations have opposite winds $\boldsymbol{v}$ and $-\boldsymbol{v}$ but equal diffusion operator ζ.

$$\frac{\partial \chi}{\partial t} + \boldsymbol{v} \cdot \boldsymbol{\nabla}\, \chi + \zeta(\chi) = \sigma \tag{3}$$

$$-\frac{\partial r}{\partial t} - \boldsymbol{v} \cdot \boldsymbol{\nabla}\, r + \zeta(r) = \pi \tag{4}$$

Diffusion is self-adjoint, $\zeta = \zeta^*$, because as the underlying turbulent motions are time symmetric, the adjoint-backward diffusion coincides with the forward diffusion.

The involvement of diffusion in backward transport is perplexing. As diffusion is irreversible, it seems to run counter the second principle. The obstacle is only apparent.

Firstly the backward integration is not intended at restoring the original position or geometry of the source. This is impossible since many sources could be an acceptable explanation for a single measurement. The backward integration just describes how the air in a sample was distributed in the atmosphere before being sampled. It is clear that far enough in the past this air was uniformly distributed.

Secondly, there is a creation of entropy towards the past by Eq. (4). This creation concerns the samples described by the π_i which are not thermodynamically closed systems. Each sample is part of the atmosphere which is an open system that permanently refreshes its organisation. The entropy of the atmosphere as a whole is roughly constant. It is maintained by the degradation of solar into thermal radiation cooled from 5880 K down to 258 K.

We stress that turbulent motions are not necessarily time symmetric. Convection for instance consists of rapid updrafts compensated by larger and slower downdrafts. Hence, the backward convection operator is obtained from the forward convection operator $\xi = \xi(\boldsymbol{x}, t)$ by means of a horizontal symmetry denoted $\xi^\dagger$. Convection is accordingly subject to the constraint $\xi^* = \xi^\dagger$ (the constraints for diffusion are stronger as $\zeta = \zeta^* = \zeta^\dagger$).

3. Rebuilding complex sources

The source σ to be identified is subject to the following constraints:

$$\mu_i = \int_{\Omega\times\mathrm{T}} \rho\, r_i\, \sigma(\boldsymbol{x}, t)\, d\boldsymbol{x}\, dt \qquad i = 1, \ldots, n$$
$$= (\sigma, r_i) \tag{5}$$

In this section we shall denote the measurement product simply as $\mu(\phi, \psi) = (\phi, \psi)$ with an associated norm $\|\phi\| = \sqrt{(\phi, \phi)}$. The infinite dimensional vector space of finitely normed functions will be denoted $\mathcal{L}^2(\Omega \times \mathrm{T})$.

The inversion of this linear system with respect to σ is under-determined. In fact all we can obtain out of the system of Eq. (5) is an estimation of the true source σ.

Measuring the μ_i amounts to determine the orthogonal projection $\sigma_\parallel$ of s on the subspace $\mathcal{D}_\parallel$ spanned in $\mathcal{L}^2(\Omega \times \mathrm{T})$ by the r_i. The very calculation of $\sigma_\parallel$ from the μ_i and r_i will be described below. Let $\mathcal{D}_\perp$ be the subspace of $\mathcal{L}^2(\Omega \times \mathrm{T})$ orthogonal to all the r_i. We recall that $\mathcal{L}^2(\Omega \times \mathrm{T})$ may be decomposed as a sum $\mathcal{L}^2(\Omega \times \mathrm{T}) = \mathcal{D}_\parallel \oplus \mathcal{D}_\perp$ with accordingly a unique decomposition $\sigma = \sigma_\parallel + \sigma_\perp$.

For the following reasons $\sigma_\parallel$ could be a good estimation of σ.
1) As $(\sigma_\perp, r_i) = 0, \quad i = 1, \ldots, n$, the measurements contain no information about $\sigma_\perp$.
2) Another estimation $z = z_\parallel + z_\perp$ obeying Eq. (5) would satisfy as well $z_\parallel = \sigma_\parallel$, hence $\|z\|^2 = \|\sigma_\parallel\|^2 + \|z_\perp\|^2 \geq \|\sigma_\parallel\|^2$. $\sigma_\parallel$ is the least norm estimation.
3) Additional measurements enlarge the space $\mathcal{D}_\parallel$ and $\|\sigma_\parallel\|^2$ grows closer to $\|\sigma\|^2$. The quality of the estimation is improved by any additional information.

The coefficients of the estimated source $\sigma_\parallel$ with respect to the r_i are obtained from the measurements after inversion of the Gramm covariance matrix H of the r_i:

$$\sigma_\parallel = \sum_{i=1}^{n} \lambda_i \, r_i \qquad \boldsymbol{\mu} = \begin{bmatrix} \mu_1 \\ \vdots \\ \mu_n \end{bmatrix} \qquad \boldsymbol{\lambda} = \begin{bmatrix} \lambda_1 \\ \vdots \\ \lambda_n \end{bmatrix} \tag{6}$$

$$H = [h_{i,j}] \quad h_{i,j} = (r_i, r_j) \quad i = 1, .., n \quad j = 1, .., n \tag{7}$$

$$\boldsymbol{\mu} = H\boldsymbol{\lambda} \quad \text{or} \quad \boldsymbol{\lambda} = H^{-1}\boldsymbol{\mu} \tag{8}$$

Physical reasons may prescribe that σ is non-zero only on a limited subdomain $\mathcal{A} \subset \Omega \times \mathrm{T}$. For instance carbon dioxide in essentially released or absorbed at the surface of the oceans or continents. ETEX source has been investigated here as such a surface source. Changing the integration domain in Eq. (5) would interfere with the definition of the scalar product. We prefer to handle the additional information by substituting for the retroplume r_i adequate restrictions $r_i|_\mathcal{A}$ in the definition and

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calculation of $\mathcal{D}_{\|}$, $\sigma_{\|}$ and H, $h_{i,j} = (r_i|_{\mathcal{A}}, r_j|_{\mathcal{A}})$.

The following notations (9), (11) and definition (10) will be convenient for the continuation of this paper:

$$\boldsymbol{r}(\boldsymbol{x},t) = \begin{bmatrix} r_1(\boldsymbol{x},t) \\ \vdots \\ r_n(\boldsymbol{x},t) \end{bmatrix} \in \mathbb{R}^n_+ \tag{9}$$

$$\boldsymbol{g}(\boldsymbol{x},t) = \begin{bmatrix} g_1(\boldsymbol{x},t) \\ \vdots \\ g_n(\boldsymbol{x},t) \end{bmatrix} = H^{-1}\,\boldsymbol{r} \in \mathbb{R}^n \tag{10}$$

$$\text{for} \quad \boldsymbol{a} \in \mathbb{R}^n, \quad \boldsymbol{b} \in \mathbb{R}^n \qquad \boldsymbol{a}\cdot\boldsymbol{b} = \sum_{i=1}^{n} a_i\, b_i \tag{11}$$

The families of functions $\{r_1, .., r_n\}$ and $\{g_1, .., g_n\}$ are dual with the meaning that (δ is Kronecker's symbol):

$$(r_i, g_j) = \int_{\Omega\times\mathrm{T}} \rho\, r_i\, g_j\, d\boldsymbol{x}\, dt = \delta_{i,j} \tag{12}$$

We write simply with these notations:

$$\sigma_{\|}(\boldsymbol{x},t) = \boldsymbol{\lambda}\cdot\boldsymbol{r}(\boldsymbol{x},t) = \boldsymbol{\mu}\cdot\boldsymbol{g}(\boldsymbol{x},t) \tag{13}$$

4. Smoothing the visibility of the network

The sources estimated according to the previous section are not satisfactory. As shown by Fig. 2 they display important positive or negative values around the position, in space and time, of the detectors. Throughout many calculations about ETEX1, considering various selections of several tenth of measurements, the conditioning of the Gramm covariance matrix (7), i.e. the ratio between the largest and smallest eigenvalues, has always been less than 20. This implies that the problem does not resort to any regularisation technique. The unsatisfactory behaviour could be expected because each retroplume displays a sharp peak concentrated around the corresponding detector. When the retroplumes are combined to estimate the source, these peaks, all at different positions, cannot compensate. From a strict mathematical point of view, the above constructions are not sound. In most practical situations the retroplumes are not modelled as elements of $\mathcal{L}^2(\Omega \times \mathrm{T})$. For instance a point detector at the origin with no wind, constant density ρ_0 and Fickian eddy diffusivity κ, is tied to a normalised retroplume with (see formula 17.50 of Seinfeld and Pandis, 1998):

$$r(l,t) = \frac{\exp\left(\frac{l^2}{4\kappa t}\right)}{(-4\pi\kappa t)^{\frac{3}{2}}} \qquad l \geq 0\,, \quad t \leq 0$$

$$\int_{-\infty}^{0}\int_{0}^{+\infty} \rho_0\, r(l,t)^2\, 4\pi l^2\, dl\, dt = +\infty \tag{14}$$

Accordingly $\|r\|^2$ does not exist and the retroplume r cannot enter in the composition of any Gramm covariance matrix. In a numerical model the steepness of the detector and of the retroplume is bounded by the size of the meshes and of the time step. The mathematical obstacle remains hidden by this uncontrolled spontaneous smoothing.

We propose to smooth the retroplumes by introducing a function $E(\boldsymbol{x},t)$, that we call “éclairement” in French or “illumination” in English, and by rewriting the measurements in the form of:

$$\mu_i = \int_{\Omega\times\mathrm{T}} \rho\, E\, r_i{}'\, \sigma(\boldsymbol{x},t)\, d\boldsymbol{x}\, dt \qquad i = 1,\ldots,n$$

$$r_i{}'(\boldsymbol{x},t) = \frac{r_i(\boldsymbol{x},t)}{E(\boldsymbol{x},t)} \tag{15}$$

This transformation amounts to using a new scalar product $(\phi,\psi)'$ (and norm $\|\psi\|'$) to represent the measurement:

$$(\phi,\psi)' = \int_{\Omega\times T} \rho\, E\, \phi(\boldsymbol{x},t)\, \psi(\boldsymbol{x},t)\, d\boldsymbol{x}\, dt$$
$$\mu_i = \left(\sigma, {r_i}'\right)' \qquad i = 1,\ldots,n \tag{16}$$

The illumination will insure that the functions ${r_i}'$ have a finite norm $\|{r_i}'\|'$ so that the Gramm covariance matrix H' might be correctly defined leading to a sound estimation:

$$H' = [h'_{i,j}] \quad h'_{i,j} = \left({r_i}',{r_j}'\right)' = \int_{\Omega\times T} \rho\, \frac{r_i\, r_j}{E}\, d\boldsymbol{x}\, dt$$
$$\boldsymbol{\lambda}' = H'^{-1}\,\boldsymbol{\mu} \qquad \sigma'_{\|}(\boldsymbol{x},t) = \boldsymbol{\lambda}'\cdot\boldsymbol{r}'(\boldsymbol{x},t) \tag{17}$$

We now describe the function E used here with a tentative and preliminary interpretation of its efficiency. These definition and interpretation are illustrated by Fig. 3.

The definition of the illumination is given with tildes symbolising precautions explained later that must be taken in order to ensure the existence of a Gramm covariance matrix against the aforementioned obstacles. The vectors $\boldsymbol{r}$, $\boldsymbol{g}$ have been defined by Eqs. (9) and (10):

$$\begin{aligned}\tilde{E}(\boldsymbol{x},t) &= {}^t\tilde{\boldsymbol{r}}(\boldsymbol{x},t)\, \tilde{H}^{-1}\, \tilde{\boldsymbol{r}}(\boldsymbol{x},t)\\ &= {}^t\tilde{\boldsymbol{g}}(\boldsymbol{x},t)\cdot\tilde{\boldsymbol{r}}(\boldsymbol{x},t) \geq 0\end{aligned} \tag{18}$$

The illumination is non-negative because the matrix $\tilde{H}^{-1}$ is positive definite. We obtain out of Eqs. (12):

$$\int_{\Omega\times T} \rho\, \tilde{E}(\boldsymbol{x},t)\, d\boldsymbol{x}\, dt = n \tag{19}$$

Note also that $\tilde{E}(\boldsymbol{x},t) = 0$ at a space-time position $(\boldsymbol{x},t)$ only if no retroplume at all goes there. On the contrary, if $\tilde{\boldsymbol{r}}(\boldsymbol{x}$,t) becomes very large in the neighbourhood of $(\boldsymbol{x},t)$,

$\tilde{E}(\boldsymbol{x},t)$ should also become very large as the matrix $\tilde{H}^{-1}$ in Eq. (18) is not position dependent. We think that $\tilde{E}(\boldsymbol{x},t)$ describes the share that goes to the neighbourhood of $(\boldsymbol{x},t)$ of the n available pieces of information produced by the measurements. Thus, $\tilde{E}$ would be an amount of information per unit mass of air (because of ρ in Eq. 19) and per unit time. This share depends on the scalar product that is used and is homogenised when changing it through the procedure (15).

The precaution should be taken for writing the above formulae to use only a square summable approximation $\tilde{\boldsymbol{r}}$ of $\boldsymbol{r}$. For instance $\tilde{\boldsymbol{r}}$ might correspond to retroplumes calculated by a numerical model. The question then arises, when $\tilde{\boldsymbol{r}}$ tends to $\boldsymbol{r}$, of what happens to $\tilde{E} = {}^t\tilde{\boldsymbol{g}} \cdot \tilde{\boldsymbol{r}}$, to the $\tilde{\boldsymbol{r}}' = \frac{\tilde{r}}{\tilde{E}}$ and finally to $\tilde{\sigma}'_{\|}$. We did not investigate this point deeply but can state our considered opinion that these variables tend to physically relevant limits E, $\boldsymbol{r}'$ and $\sigma'_{\|}$. Suppose some r_i has a non square summable singularity for a position $(\boldsymbol{x}_0,t_0)$ such as described by Eqs. (14). As r_i describes the dispersion of a finite amount of inverse tracer the singularity remains simply summable. Accordingly when $\tilde{r}_i$ tends to r_i the integral Eqs. (12), $\int_{\Omega\times\mathrm{T}} \rho\, \tilde{r}_i\, \tilde{g}_j\, d\boldsymbol{x}\, dt = \delta_{i,j}$, may be maintained with minor changes of the $\tilde{g}_j$; in particular there is no requirement that $\tilde{g}_j(\boldsymbol{x}_0,t_0) \rightarrow 0$ in order to compensate for $\tilde{r}_i(\boldsymbol{x}_0,t_0) \rightarrow +\infty$. We also think that $\tilde{E}$ will tend to a non negative illumination E subject to the constraint (19) and having a simply summable singularity at $(\boldsymbol{x}_0,t_0)$ in such a way that the following norm now derives from an integral: $\|\sigma'_{\|}\|' = \left(\int \rho\, E\, {\sigma'_{\|}}^2\, d\boldsymbol{x}\, dt\right)^{\frac{1}{2}}$.

The calculation of the estimated source $\sigma'_{\|}$ will require the calculation and inversion of a first Gramm covariance matrix H to obtain E, and of a second smoothed Gramm covariance matrix H' to finally obtain $\sigma'_{\|}$. In the calculations the conditioning of H', calculated from smoothed retroplumes, was always a bit less favourable than that of H. It never exceeded 60. Given that, as shown in Sect. 6, it is the square root of the conditioning that counts, regularisation techniques are unessential at this stage.

Note that after smoothing an equation analogous to (19) may be written from which is inferred the smoothed share of information E':

$$\int_{\Omega\times T} \rho\, E\, {}^t\boldsymbol{r}'(\boldsymbol{x},t)\, H'^{-1}\, \boldsymbol{r}'(\boldsymbol{x},t)\, d\boldsymbol{x}\, dt = n$$
$$E'(\boldsymbol{x},t) = E(\boldsymbol{x},t)\, {}^t\boldsymbol{r}'(\boldsymbol{x},t)\, H'^{-1}\, \boldsymbol{r}'(\boldsymbol{x},t) \tag{20}$$

In the calculations described by Sects. 7, 8, for Eqs. (15), (16), (17), (20), the lowest values of the illumination were bounded not to depart more than a factor 1000 from the maximum value. On the one hand this is sufficient to cancel the spots of illumination rising from the background with this factor 1000 for horizontal dimensions of about 100 km, stretched by the time integration on Fig. 3a1, b1, unreasonably small compared to the distances between neighbouring stations. On the other hand this avoids to give an excessive importance to badly seen regions where $\boldsymbol{r}$ is little; as E varies like $\|\boldsymbol{r}\|^2$, $\boldsymbol{r}'$ would be as large as $\|\boldsymbol{r}\|^{-1}$. The resulting smoothed illumination is shown on Fig. 3a2, b2.

In the continuation of this document the $'$ will no longer be written as an indication for the use of the above smoothing technique.

5. Rebuilding non-negative complex sources

There are physical circumstances in which the source $\sigma(\boldsymbol{x},t)$ is known to be non-negative and should be attributed a non-negative estimation. This will be the case when considering such anthropic pollutants as methane, carbon monoxide, pesticides, etc. We recall that the linear decay processes should be considered a transport term in the l.h.s. of Eq. (3) rather than a negative contribution to the r.h.s. source σ. There is no reason why the algebraic estimation $\sigma_{\|}$ built as a linear combination of the retroplumes should be non-negative. The algebraic and positive estimation of the real source may

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be given the following definitions:

$$\sigma_{\parallel} = \mathrm{argmin}\,\{\ \|s\|,\ (s, r_i) = \mu_i\,,\ i = 1, .., n\ \} \tag{21}$$

$$\sigma^{+} = \mathrm{argmin}\,\{\ \|s\|,\ (s, r_i) = \mu_i\,,\ i = 1, .., n,\ s \geq 0\ \} \tag{22}$$

We have taken the non-negative constraint into account in a computationally cost-effective manner. The calculation is based upon the property (29) that can be controlled only at the end and was always controlled to be true as explained below. Starting with $\sigma_{\parallel}$ we obtain at iteration ν a source σ^{ν} constrained by the measurements and additionally constrained to be zero wherever $\sigma^{\nu-1}$ was negative or zero. The additional accumulative constraints are easily handled by just restricting the integral calculation of the Gramm covariance matrix (Eq. 25). The limit of the procedure is a source σ^{∞} constrained by the measurements and everywhere positive or zero. Numerically we obtained it in six to twenty two iterations. There is no obvious reason why σ^{∞} and σ^{+} should coincide. Nevertheless when controlling the optimality conditions we always concluded that, to a surprising approximation, they do. We have to mention that the calculations were smoothed and regularised as described in Sects. 4 and 6.

Let's describe more technically the procedure used in this paper. Sect. 3 is taken to be iteration number 0 with $\Omega \times \mathrm{T}$, H, $\sigma_{\parallel}$ now termed $\mathcal{O}^0_+$, H^1, σ^1; $\mathcal{O}^0_-$ is the empty set. Then:

$$\begin{cases} \sigma^{\nu} = \mathrm{argmin}\left\{\|s\|,\ (s, r_i) = \mu_i\,,\ i = 1, .., n,\ s|_{\mathcal{O}^{\nu-1}_-} = 0\right\} \\ \mathcal{O}^{\nu}_- = \Omega \times \mathrm{T} - \mathcal{O}^{\nu}_+ = \left\{(\boldsymbol{x}, \mathrm{t})\ /\ \sigma^{\nu-1}(\boldsymbol{x}, \mathrm{t}) \leq 0\right\} \end{cases} \tag{23}$$

Note that: $\mathcal{O}^1_- \subset \mathcal{O}^2_- \subset \ldots$ and $\mathcal{O}^1_+ \supset \mathcal{O}^2_+ \supset \ldots$. The set where $\sigma^{\nu-1}$ is negative or zero is $\mathcal{O}^{\nu}_-$. There, σ^{ν} is constrained to vanish, and also by induction the limit σ^{∞}. Equation (2) may accordingly be replaced by:

$$\mu_i = \int_{\mathcal{O}^{\nu-1}_+} \rho(\boldsymbol{x}, t)\ r_i(\boldsymbol{x}, t)\ \sigma^{\nu}(\boldsymbol{x}, t)\ d\boldsymbol{x}\ dt \tag{24}$$

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σ^ν is obtained after inverting the Gramm covariance matrix:

$$H^\nu = [h^\nu_{i,j}] \qquad h^\nu_{i,j} = \int_{\mathcal{O}^{\nu-1}_+} \rho\, r_i\, r_j\, d\boldsymbol{x}\, dt \tag{25}$$

$$\boldsymbol{\lambda}^\nu = (H^\nu)^{-1}\boldsymbol{\mu} \qquad \sigma^\nu(\boldsymbol{x},t) = \begin{cases} \boldsymbol{\lambda}^\nu \cdot \boldsymbol{r}(\boldsymbol{x},t) & \text{in } \mathcal{O}^\nu_+ \\ 0 & \text{in } \mathcal{O}^\nu_- \end{cases} \tag{26}$$

and in the limit:

$$\begin{array}{l} \boldsymbol{\lambda}^\nu \to \boldsymbol{\lambda}^\infty \\ \mathcal{O}^\nu_- \to \mathcal{O}^\infty_- \end{array} \qquad \sigma^\infty(\boldsymbol{x},t) = \begin{cases} \boldsymbol{\lambda}^\infty \cdot \boldsymbol{r}(\boldsymbol{x},t) & \text{in } \mathcal{O}^\infty_+ \\ 0 & \text{in } \mathcal{O}^\infty_- \end{cases} \tag{27}$$

In order to decide whether σ^∞ is indeed σ^+ we just have to control that the linear combination $\boldsymbol{\lambda}^\infty \cdot \boldsymbol{r}$ is negative or zero wherever in $\mathcal{O}^\infty_-$. Indeed one first notices that $-2\lambda^\nu_i, \ldots, -2\lambda^\nu_n$ are the Lagrange multipliers of σ^ν for the measurement constraints $\int_{\Omega\times T} \rho\ r_i\ \sigma^\nu\ d\boldsymbol{x}\ dt - \mu_i\ = 0$. Hence (see for instance the presentation by Culioli, 1994), for all $(\boldsymbol{x},t)$ in $\mathcal{O}^\infty_-$, by showing $\boldsymbol{\lambda}^\infty \cdot \boldsymbol{r}(\boldsymbol{x},t) < 0$ we would prove the zero equality constraint at $(\boldsymbol{x},t)$ to be active and equivalent to a positivity constraint. For all the calculations performed here we have obtained the following inequality indicating that σ^∞ is at least a very good approximation of σ^+. The first part of the inequality is the amount of tracer positively emitted by $\boldsymbol{\lambda}^\infty \cdot \boldsymbol{r}$ in $\mathcal{O}^\infty_-$ thus violating the optimality conditions. The second part is the amount of tracer released by σ^∞.

$$\int_{\mathcal{O}^\infty_-} \rho\ \max\left(0, \boldsymbol{\lambda}^\infty \cdot \boldsymbol{r}(\boldsymbol{x},t)\right)\ d\boldsymbol{x}\ dt \le\ 3\ 10^{-2} \int_{\mathcal{O}^\infty_+} \rho\ \sigma^\infty(\boldsymbol{x},t)\ d\boldsymbol{x}\ dt \tag{28}$$

We tried to improve the optimality of σ^∞ by relaxing the violated optimality conditions. This means that we went once again through the iterative system (23) with a new set

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$\mathcal{O}_+^0$ defined as the former set $\mathcal{O}_+^\infty$ plus all parts of $\mathcal{O}_-^\infty$ where the optimality conditions were violated. The inequality (28) could not be improved significantly.

The equality $\sigma^\infty = \sigma^+$ is equivalent to:

$$\sigma_\parallel(\boldsymbol{x}, t) \le 0 \Longrightarrow \sigma^+(\boldsymbol{x}, t) = 0, \quad (\boldsymbol{x}, t) \in \Omega \times \mathrm{T} \tag{29}$$

We suspected, but could not prove, this implication to be true in such quadratic optimal problems as (21), (22) whenever $r_i \ge 0$ and $\mu_i \ge 0$. A similar conjecture has already been proposed, based upon computational results, in the conclusions of de Villiers, McNally and Pike (1999). The following counter-example is due to Baillon (2003) who kindly communicated it to us. It implies 4-dimensional "retroplumes" $r_1 = (2, 5, 4, 4)$ $r_2 = (1, 6, 9, 4)$ $r_3 = (7, 8, 3, 3)$; the constraints for $s \in \mathbb{R}^4$ are $r_i \cdot s = 1,\ i = 1, 2, 3$. The orthogonal projection is $\sigma_\parallel = \left(-\frac{307}{23982}, \frac{904}{11991}, -\frac{141}{7994}, \frac{308}{1713}\right)$, the positive optimum is $\sigma^+ = \left(\frac{1}{39}, \frac{1}{39}, 0, \frac{8}{39}\right)$.

We think the above algorithm is interesting because it is cost effective and respects the logic of the physical problem which is important when regularising techniques have to be used. Rather than controlling and improving the optimality conditions at the end of the calculation, of course it would be more satisfactory understanding a possible link between the retroplumes and the property (29).

6. Noises and regularisation

The use of regularisation techniques, not essential for calculating the algebraic estimation of the source, becomes such when considering a positive estimation. Indeed the iterative removal of parts of the domain $\Omega \times \mathrm{T}$ corresponding to significant contrasts between the retroplumes rapidly degrades the conditioning of the Gramm covariance matrix, already at least 300 for H^2. This point is addressed in the present section which furthermore investigates the effect of noises on the quality of the estimations.

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By looking at Eqs. (2), (17) or (24) we see that the calculation of $\boldsymbol{\lambda}$ (or $\boldsymbol{\lambda}^{\nu}$) is touched firstly through the vector $\boldsymbol{\mu}$ by the quality of the measurements and secondly through the Gramm covariance matrix H (or H^{ν}) by the quality of the transport data. The effect of these errors may be controlled by just improving the conditioning of H before inversion. Let's denote as μ_i^o the value effectively observed for the i^{th} measurement displaying a noise gap $\delta\mu_i$ from the theoretical value μ_i. The estimated coefficients λ_i^o and source $\sigma_{\parallel}^o(\boldsymbol{x},t)$ will correspondingly display errors $\delta\lambda_i$ and $\delta\sigma_{\parallel}(\boldsymbol{x},t)$ thus drifting from the theoretical values.

$$\mu_i^o = \mu_i + \delta\mu_i \qquad \mu_i = \int_{\Omega\times\mathrm{T}} \rho\, r_i\, \sigma\, d\boldsymbol{x}\, dt \tag{30}$$

$$\lambda_i^o = \lambda_i + \delta\lambda_i \qquad \sigma_{\parallel}^o = \sigma_{\parallel} + \delta\sigma_{\parallel} \tag{31}$$

Now suppose simply that the same measurement has been performed twice by two identical detectors operated simultaneously and at the same place. The observations $\mu_1^o = \mu + \delta\mu_1$ and $\mu_2^o = \mu + \delta\mu_2$ correspond to the same theoretical measurement $\mu = \mu_1 = \mu_2$ and retroplume $r = r_1 = r_2$. The Eqs.(8) may now be written as:

$$\begin{bmatrix} \frac{\mu_1^o+\mu_2^o}{2} \\ \frac{\mu_1^o-\mu_2^o}{2} \end{bmatrix} = \begin{bmatrix} \|r\|^2 & 0 \\ 0 & 0 \end{bmatrix} \begin{bmatrix} \frac{\lambda_1^o+\lambda_2^o}{2} \\ \frac{\lambda_1^o-\lambda_2^o}{2} \end{bmatrix}$$

$$\begin{bmatrix} \frac{\lambda_1^o+\lambda_2^o}{2} \\ \frac{\lambda_1^o-\lambda_2^o}{2} \end{bmatrix} = \begin{bmatrix} \|r\|^{-2} & 0 \\ 0 & +\infty \end{bmatrix} \begin{bmatrix} \mu + \frac{\delta\mu_1+\delta\mu_2}{2} \\ \frac{\delta\mu_1-\delta\mu_2}{2} \end{bmatrix} \tag{32}$$

This example shows that the two pieces of information μ_1^o, μ_2^o may be rearranged and divided into a 'relevant' part $\frac{\mu_1^o+\mu_2^o}{2}$ and a 'redundant' part $\frac{\mu_1^o-\mu_2^o}{2}$. The redundant information corresponds to non significant differences between repeated (combinations of) measurements with the risk of an abusive interpretation in terms of the noise. It jeopardises with little eigenvalues the conditioning of the Gramm covariance matrix. The

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presence of redundant information is not totally negative as it reduces the noise touching the relevant information. If the noises in μ_1^o and μ_2^o are independent with the same variance (the bar denotes the statistical expectation), $a = \overline{\delta\mu_1{}^2} = \overline{\delta\mu_2{}^2}$ and standard deviation $s = \sqrt{a}$, the effective noise for the relevant information $\frac{\mu_1^o+\mu_2^o}{2}$ has a standard deviation $s' = \frac{s}{\sqrt{2}}$.

In order to improve the quality of the relevant information and to remove the redundant information we regularised the system (2) (or rather 24) by a method called Truncated Singular Value Decomposition (TSVD). An extensive description can be found in (Bertero, de Mol and Pike, 1985, 1988). H is put into diagonal form; as it is positive definite, the transition matrix O is orthogonal (${}^tO = O^{-1}$, the left exponent t stands for the transposition). The eigenvalues written in decreasing order display a conditioning $\frac{d_1}{d_n}$ that will be reduced to the desired value $cond$ by topping the inversion of the too little ones:

$$H = O \begin{bmatrix} d_1 & & & & \\ & \ddots & & & \\ & & d_k & & \\ & & & d_{k+1} & \\ & & & & \ddots \\ & & & & & d_n \end{bmatrix} O^{-1} \tag{33}$$

$$d_1 \geq .. \geq d_k \geq \kappa > d_{k+1} \geq .. \geq d_n \geq 0 \qquad \kappa = \frac{d_1}{cond}$$

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The regularised inverse of H is taken to be:

$$H^{inv} = O \begin{bmatrix} d_1^{-1} & & & & \\ & \ddots & & & \\ & & d_k^{-1} & & \\ & & & \kappa^{-1} & \\ & & & & \ddots \\ & & & & & \kappa^{-1} \end{bmatrix} O^{-1} \tag{34}$$

The theoretical regularised estimation of the source will be now:

$$\sigma_{\|}(\boldsymbol{x},t) = \boldsymbol{\lambda} \cdot \boldsymbol{r}(\boldsymbol{x},t) \qquad \boldsymbol{\lambda} = H^{inv} \boldsymbol{\mu} \tag{35}$$

When using the observations $\boldsymbol{\mu}^o = \boldsymbol{\mu} + \boldsymbol{\delta\mu}$ this estimation is realised with some error as:

$$\sigma_{\|}^o = \sigma_{\|} + \delta\sigma_{\|} = \boldsymbol{\lambda}^o \cdot \boldsymbol{r} \qquad \boldsymbol{\lambda}^o = H^{inv} \boldsymbol{\mu}^o \tag{36}$$

$$\delta\sigma_{\|}(\boldsymbol{x},t) = \boldsymbol{\delta\lambda} \cdot \boldsymbol{r}(\boldsymbol{x},t) \qquad \boldsymbol{\delta\lambda} = H^{inv} \boldsymbol{\delta\mu} \tag{37}$$

In order to evaluate the importance of this error let's introduce the statistical covariance matrix A of the measurement errors, not to be mistaken with the Gramm covariance matrix H:

$$A = [a_{i,j}] \quad a_{i,j} = \overline{\delta\mu_i \, \delta\mu_j} \quad i = 1,..,n \quad j = 1,..,n \tag{38}$$

We now evaluate finally as the trace of a square matrix $\|\delta\sigma_{\|}\|^2 = {}^t\delta\lambda \; H \; \delta\lambda = {}^t\delta\mu \; H^{inv} \; \delta\mu = tr\left(H^{inv} \, [\delta\mu \, {}^t\delta\mu]\right)$. A statistical expectation is obtained:

$$\overline{\|\delta\sigma_{\|}\|^2} = tr(H^{inv} A) \tag{39}$$

This expression is adapted from (Hunt, 1971) with nevertheless significant differences due to the structure of the present problem. It may be used to generalise the

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conclusion that the redundant information reduces the effect of the noise but must be removed from the inversion. It shows also that when estimating the standard deviation $\sqrt{\overline{\|\delta\sigma_{\|}\|^2}}$ it is not the conditioning of H that counts but rather its square root. Indeed satisfactory results are obtained with a conditioning limit cond=60.

One should also notice that our limited knowledge of the transport has repercussions on the available retroplumes $r_i^o = r_i + \delta r_i$. This introduces in the estimation of the source a bias that could be tracked through the formulae. We preferred to evaluate the bias directly in the calculations.

7. Calculations about ETEX1

In October 1994, between 23, 16:00 UT and 24, 04:00 UT, 340 kg of an inert tracer, perfluoromethylcyclohexane or pmch, were released from a ground position near the village of Monterfil, Brittany, France. Time averaged concentration measurements were delivered each 3 h by 168 detectors all over Europe. The experiment, sponsored by the European Commission together with a second one organised in November the same year (ETEX2), is described in (Joint Res. Centre, 1998). The data are available from the Joint Research Centre, Environment Monitoring Unit, Ispra (Varese), Italy (http://java.ei.jrc.it/etex/database/).

The results presented here have been obtained from two selections of measurements described on Fig. 1. The first selection comprises 48 measurements from 7 stations making up a subnetwork with a characteristic spacing of 300 km. The second one comprises 130 measurements taken by 14 stations with a characteristic spacing of 500 km. The stations have been selected in order in order to detect the cloud of pmch and its edges. The measurements have been selected for each station in order to capture the arrival, passing and the end of the cloud. In such a station as Brest (station number 89) we could only take into account zero valued measurements containing the information that the cloud had never been there.

In order to investigate the effect of detector and meteorological errors, the source ETEX1 has been rebuilt for different values of the selected measurements:
1) synthetic measurements obtained from POLAIR $\mu_1^s, .., \mu_n^s$
2) synthetic measurements with a Gaussian noise $\mu_1^s(1+\alpha_1), .., \mu_n^s(1+\alpha_n)$, with $\overline{\alpha_i} = 0$, $\sqrt{\overline{\alpha_i^2}} = 30\%$
3) real measurements from the ETEX1 database $\mu_1^o, .., \mu_n^o$

In order to investigate the potential of the algorithm three artificial sources have been defined and reconstructed with synthetic values for the selection of 130 measurements. These sources correspond to a 12 h release of 340 kg of tracer:
1) in southern Belgium, 50°00'N, 4°30'E, beginning on 23 October 1994, 16:00 UT
2) on the coast of the Netherlands, 53°00'N, 4°30'E, beginning on 24 October 1994, 16:00 UT
3) on the German-Polish boarder, 53°30'N,14°30'E, beginning on 25 October 1994, 16:00 UT

The retroplumes were obtained by means of the atmospheric transport model POLAIR (Sportisse et al., 2002; Sartelet et al., 2002) developed at the Centre d'Enseignement et de Recherche Eau, Ville, Environnement. POLAIR is the fruit of a close cooperation with the team in charge at Electricité de France of the passive atmospheric transport model Diffeul (Wendum, 1998). It is a fully modular 3D Eulerian chemistry transport model. Advection is solved with a flux limiter method; diffusion is solved by a three point scheme. The reactive part of the model was switched off for the present application. In order to cover western Europe we used a grid extending from 15°W to 35°E and from 40°N to 67°N. The horizontal resolution of the model was 0.5° × 0.5° with fourteen Cartesian levels at 32, 150, 360,..., 6000 m above ground or sea level. The calculation went in inverse mode through a period of time starting the 27 October 1994, 07:00 UT (end of the last 3 h sampling period) and finishing back

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in time the 15 October 1994, 00:00 UT. This 296 h period was covered with a 15 min time step; concentrations were stored each hour. Meteorological data produced by the European Centre for Medium Range Weather Forecast were kindly supplied by Météo France. These six-hourly data had the same horizontal resolution as POLAIR but had to be interpolated according to the vertical Cartesian levels of the model. Each retoplume required 3 mn CPU time on a PC (Xeon, 1.5 GHz, 1 Go RAM).

The sources of most tracers of interest, carbon dioxide, carbon monoxide, methane, are essentially spread at ground or sea level. This is a physical simplification that must be taken into account in order to improve the quality of the reconstruction. Hence the retroplumes have been restricted to the surface (ground or sea level) before going through the inversion.

The calculation of the algebraic and positive sources tied to a certain family of measurements was organised as follows. A first Gramm covariance matrix H_0 was calculated by visiting each of the $93*54*296 \simeq 1.5\ 10^6$ space-time meshes at ground or sea level in order to increment the coefficients $h_{i,j} = \int_{\Omega\times T} \rho\ r_i\ r_j\ d\boldsymbol{x}\ dt$. A first inversion was performed in order to calculate the "éclairement" E. The retroplumes were divided by E (with the little values of E limited by $\frac{\max E}{1000}$). A second Gramm covariance matrix H^1 was calculated out of the smoothed retroplumes leading now, through a second inversion, to the algebraic estimation of the source. Successive Gramm covariance matrices H^ν were iteratively obtained, regularised and inverted in order to reach a positive estimation of the source. The conditioning of the first two inversions was always less than the empirically chosen limit cond=60, but most of the successive inversions required to obtain a positive estimation had to be regularised. The choice $cond = 60$ provides a satisfactory stability of the calculation with respect to observational noises as can be seen by comparing Figs. 4c and 4d or 5c and 5d. Finally, we controlled the optimality conditions of this positive source. The algebraic estimation restored to within 1% of error the values of the largest measurements through five orders of magnitude. The positive estimation restored to within 10% of error the values of the largest measurements through two orders of magnitude. The fact that a measurement is negligible

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is always restored.

The calculation time for the algebraic source with 48 measurements was 2 mn CPU on a PC (Xeon, 1.5 GHz, 1 Go RAM), plus 1 more minute CPU to reach the positive source and control its optimality. With 130 measurements these times become 10 and 5 min, respectively. Most of the time is used for calculating the Gramm covariance matrices. The regularised inversion is very rapid. The calculation time would be roughly multiplied by the number of levels, 14, if the hypothesis of a superficial source was renounced. It is clear that this hypothesis is made only for its physical relevance and not for numerical convenience.

8. Results and discussion

An animation would be the best way to view sources rebuilt as functions of time at the surface (ground or sea level). As this is not suitable for a paper we decided to cancel the time by showing sources integrated in time for each position $\Sigma(\boldsymbol{x}) = \int \sigma(\boldsymbol{x}, t)\, dt$.

The reconstructions obtained from non smoothed retroplumes, shown by Fig. 2 are not realistic. The source is obtained as a collection of peaks corresponding to each measurements. This reconstruction amounts to explaining each measurement separately from the others by a local source just a bit spread by the peaked geometry of the corresponding retroplume. The local sources need not be very large so that total amount of tracer released is clearly underestimated. This unphysical reconstruction is also mainly positive, except for little negative compensations as can be seen by the English stations UK7, UK8 on Figs. 2a1 or b1. These reconstructions follow the geometry of the illumination represented by Figs. 3a1, b1. Notice that the illumination is a property of the monitoring system independently of any effective source.

When the retroplumes are smoothed as explained in Sect. 4 the reconstruction becomes more coherent. On Figs. 4a, 4b, or 5a, 5b, obtained for synthetic measurements, the maximum value of the source is obtained for the expected position of Monterfil. The quality of the reconstruction is also improved, as shown by Figs. 4c, 5c, when

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using the real data but the maximum of the source may be inaccurate, probably due to model, especially wind biases.

The relative uncertainty in the real data is about 15% (Joint Res. Centre, 1998). This is twice smaller than the 30% of noise introduced in the synthetic measurements with no significant variation of the algebraic or positive reconstructions as shown by Figs. 4a, 4b and 5a, 5b. On the contrary when using real data, as shown by Figs. 4c or 5c, significant differences occur especially in terms of the total positive or negative contributions. Hence, when studying ETEX1, the influence of the observational noise is negligible compared to the influence of model biases. These biases stem in particular from the limited quality of the analysed winds and of the diffusion parametrised according to Louis (1979) and Louis et al. (1982). The line of current departing from the release ETEX1, first flowing eastward progressively bends northward through Germany before passing in Scandinavia. It seems this current line went more east in the model than in the reality. For that reason the concentration of pmch really observed in the Danish station DK5 is more than can be modelled in POLAIR from the source ETEX1. During the inversion of the 130 real measurements, the “excessive” values of the measurements are seen like a local source above the station DK5: compare Figs. 5a and 5c. Stations D32, DK4 and PI6 display an opposite behaviour.

In fact, wind or model biases, when confronted to real measurements, generate artificial positive or negative releases displaying extrema by the position, in space and time, of the detectors. Such artificial extrema coinciding with the position of the detectors are easily identified even if the effective source of tracer is not known. They provide thus a valuable information about the biases of the model, especially the biases of the windfield. To a lesser extent in the case of ETEX1 such extrema are also generated by the observational noise. They could also correspond to the reality if the source of tracer is very inhomogeneous and if a detector has been settled close to an important point of release; this explanation is nevertheless not suitable for negative extrema.

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The comparison of the results obtained from the two selections is surprising. In order to avoid any interference of noises or biases the comparison should be mainly between Figs. 4a and 5a. The algebraic reconstruction out of 48 measurements looks poorer than that out of 130 measurements, but the positive reconstruction is on the contrary much better. This conclusion is valid for both the geographic distribution of the sources and the total positive or negative contributions. We think that the 48 measurement selection provides a better observation of the source ETEX1. This is due to the presence, in this selection only, of measurements taken close to Monterfil by the station F2. The algebraic reconstruction out of 48 measurements turns out poorer but in fact it is more complex because it is tied to a richer information. The quality of the positive reconstructions on Figs. 4a2 and 5a2 displays a better agreement with the quality of the data. Our interpretation is that the algorithm is mathematically sound but some physical constraint, a positivity constraint for instance, is required in order to obtain a physically sound result. Positivity is not an acceptable constraint for carbon dioxide. It is generally considered that smoothness is an important feature of this source; such gradients as represented on Fig. 4a1 would not be acceptable for it. Hence a future evolution of the algorithms described in this paper should be the definition and implementation of some smoothness constraint. This new problem of smoothness of course should not be mistaken for the one addressed in Sect. 4. We must also mention that the source ETEX1 investigated here is all but smooth. The case of smooth sources should now be investigated as such. It seems that a source varying with typical distances of some hundreds of kilometers would be well seen by such a network as the ETEX subnetworks used in this paper. This resolutions could probably be even better if the time variations of the source are slow too.

The word quality used above to describe the relevance of a set of measurements with respect to a source may be given an accurate meaning through the concept of illumination. As shown by Fig. 3 the illumination of the source ETEX1 is better with the 48 measurements than with the 130 ones. In order to confirm

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this additional interpretation of the illumination we evaluated synthetic values for the selection of 130 measurements tied to artificial sources at various positions and dates. Then the sources were reconstructed. Two sources in Belgium and north eastern Germany have been chosen in order to receive the same high illumination a third one was chosen in the Netherlands with a ten times lower value similar to that received by ETEX1. The results reported on Fig. 6 are very explicit. The first two sources are reconstructed with a very good accuracy, the third one is reconstructed with the same accuracy as ETEX1 on Fig. 5a.

Acknowledgements. The author is deeply grateful to J.-B. Baillon controlling a conjecture, to R. Cominetti explaining the subtlety of optimisation theory under constraints, to A. Osses pointing out possible links with Carleman's control techniques, to L. Gallardo Klenner and B. Sportisse organising these contacts, to B. Cabrit and to J. Baverel for fruitful past discussions. Graphics have been prepared with the user friendly and public domain graphical package named GrADS originally developed by B. Dotty (COLA, support@grads.iges.org) and maintained with the help of M. Fiorino (LLNL).

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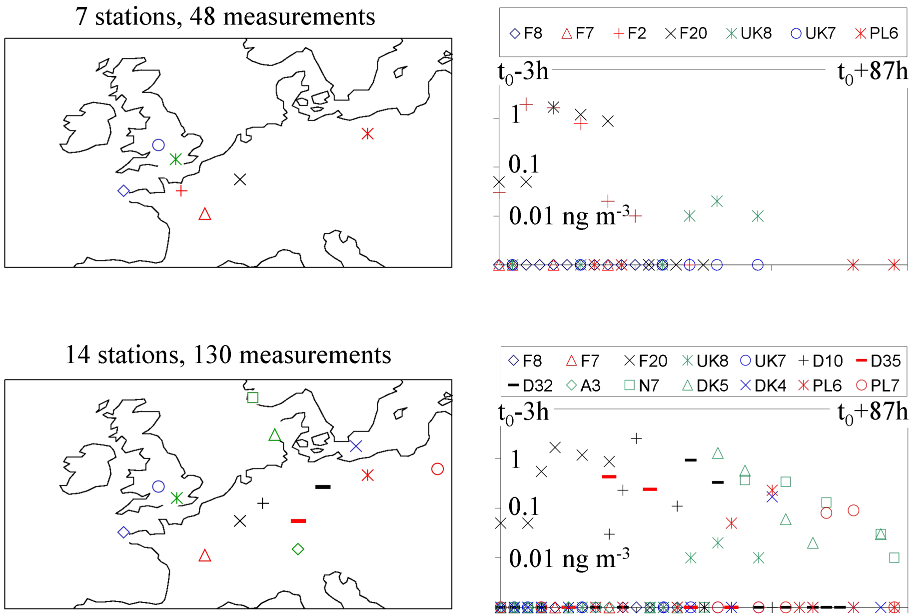

Fig. 1. Geographic and time distribution of the two selections of measurements used for the calculations. Upper two images, the 48 measurement selection, lower two images the 130 measurement selection. The time t_0 corresponds to the beginning of the pmch release in Monterfil, 23 October 1994, 16:00 UT; the first sampling period begins at $t_0 - 3h$. Different detectors are indicated by different symbols. The values in ng of pmch per m^3 are from the ETEX1 database.

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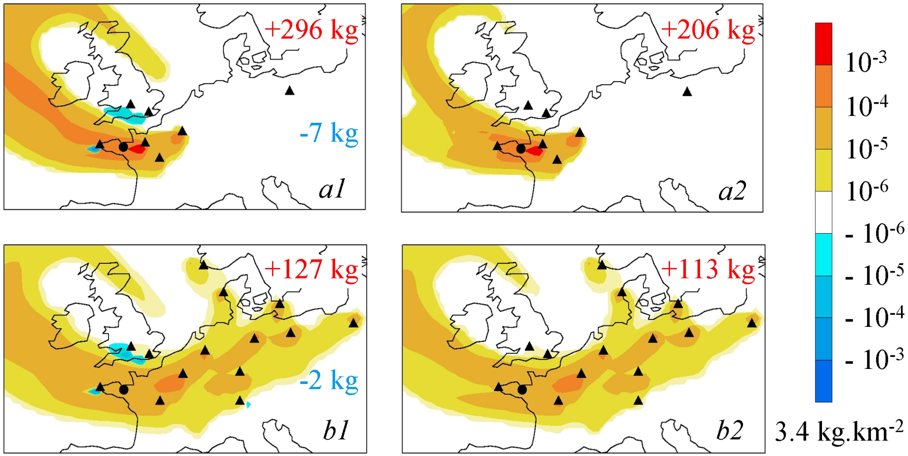

Fig. 2. Unsatisfactory results obtained out of a direct application of the orthogonal projection method for the two selections of 48 and 130 measurements in **(a)** and **(b)**, respectively. The sources, evaluated as ground or sea level fluxes out of synthetic measurements, are integrated in time for each position. The positivity constraint is taken into account on the right column. The black dot indicates the position of Monterfil, the triangles indicate the position of the selected detectors.

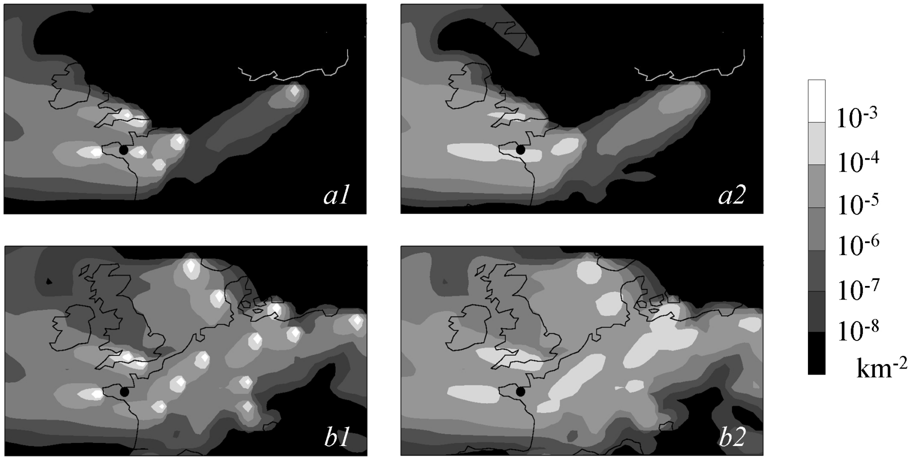

Fig. 3. Time integrated maps of "éclairement" or "illumination" for the selections of 48 measurements in **(a)** and 130 in **(b)**: initial illumination on the left, smoothed illumination on the right. The illumination is a geometric property describing, independently of the effective values of the measurements, which regions have been observed. The black parts of the maps correspond to areas poorly or not seen at all. For the detectors these areas are as if they did not exist as sources there would have no influence on the measurements. The unit, km^{-2} after the time integration and for sources known to be surface sources, may be regarded as a number of information per unit area for an integrated total of 48 or 130, respectively.

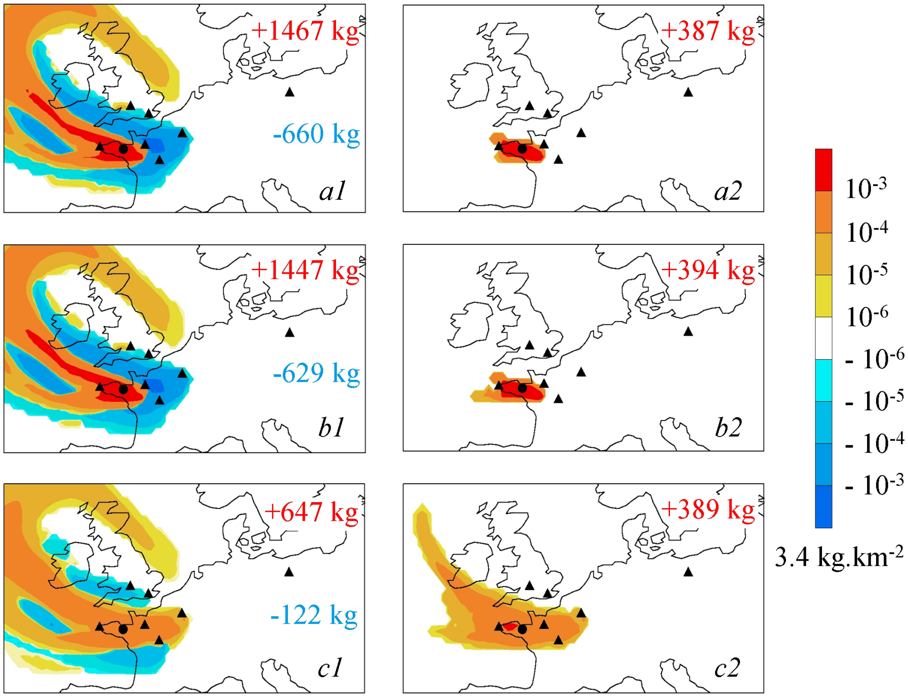

Fig. 4. Results obtained for the selection of 48 measurements when the illumination is taken into account as explained in Sect. 4. The source is evaluated as a flux at ground or sea level. Monterfil is indicated by a black point and the detectors by triangles. The part **(a)** of the figure is obtained for synthetic measurements, part **(b)** for synthetic measurements with 30% of relative noise, part **(c)** out of the real measurements from the ETEX1 database. Because of the time integration the positive reconstruction, on the right column, may sometimes be non zero at places where the algebraic reconstruction, on the left column, is negative in apparent contradiction with the proposition (29). The sum of the positive contributions is indicated in red, in blue for the negative ones.

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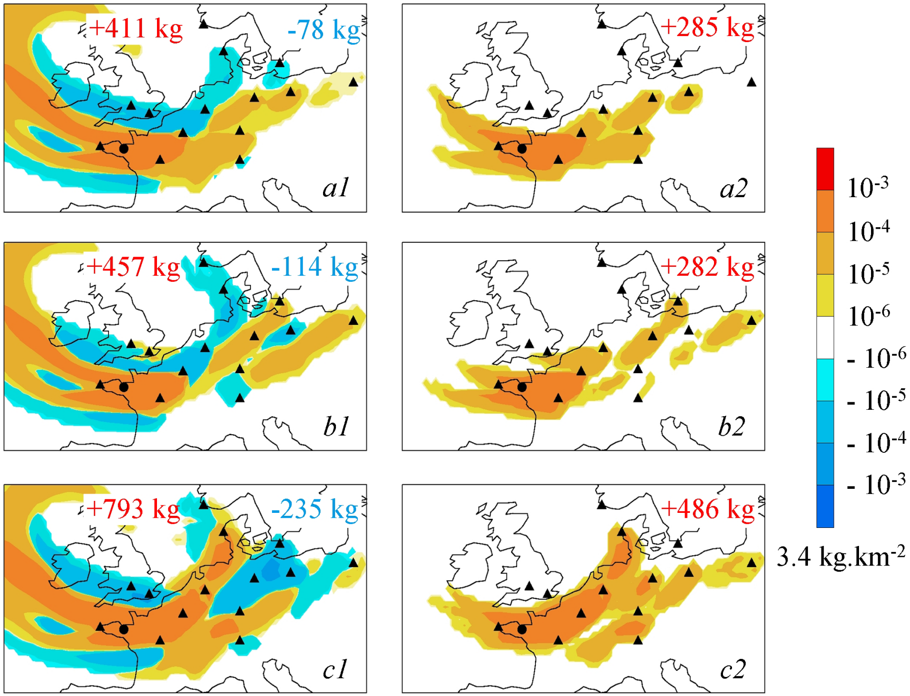

Fig. 5. Results obtained for the selection of 130 measurements. The organisation of this figure is parallel to that of Fig. 4.

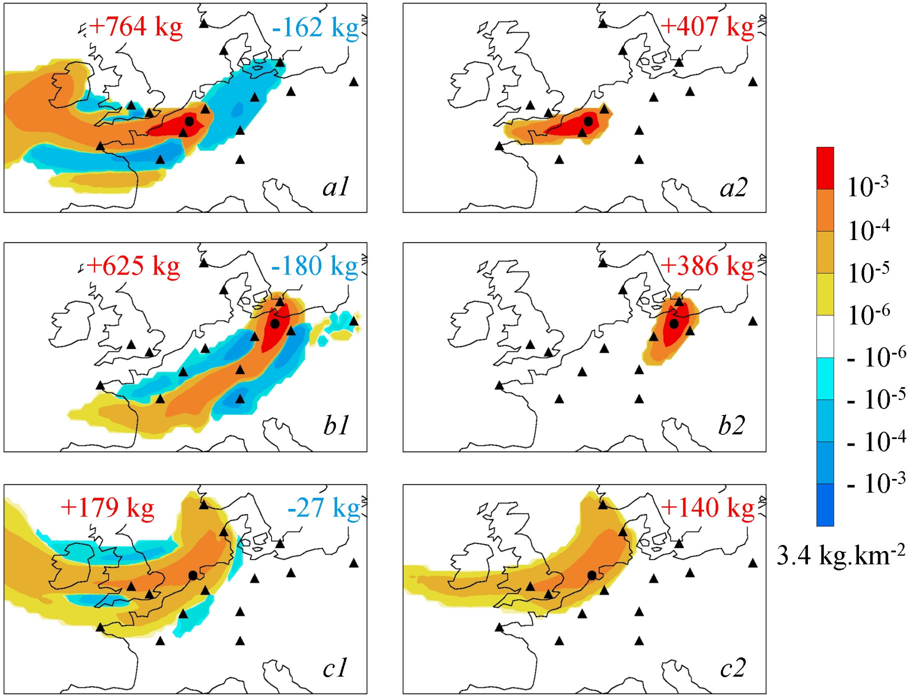

Fig. 6. Algebraic and positive reconstructions out of synthetic values of the selected 130 measurements for three imaginary sources at positions indicated by the black point: **(a)** in Belgium, **(b)** in north-eastern Germany, **(c)** in the Netherlands. The sources correspond to 340 kg of tracer released during the 12 h of best local illumination. As explained for Fig. 4 the results are integrated in time. In (a) and (b) the sources are well illuminated, the reconstruction has a good accuracy, especially in the positive case. In (c) the lesser illumination of the source in the Netherlands is similar to that of the source ETEX1 and so is the lesser quality of its reconstruction.

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